



DECLARATION

I, Takeshi TABATA, a Japanese citizen, of 16 Higashitukamotocho, Shimogamo, Sakyo-ku, Kyoto-shi, Kyoto 606-0855 JAPAN, do solemnly and sincerely declare as follows:

1. I am one of the inventors of the subject matter of U.S. Patent Application No.08/765,046 and I am familiar with the contents of this application.

2. I was born on March 1, 1962.

3. I graduated the University of Tokyo, Department of Chemistry, Faculty of Science in March, 1984.

4. I obtained Master of Science, in March, 1986 from the University of Tokyo.

5. I have been working in Osaka Gas Company Limited since 1986, and have been engaged in catalyst research at research laboratory for 10 years since 1988.

6. Job title: Manager, Catalysis and Sensors, Fundamental Research Laboratories

Degree: Doctor of Engineering from the University of Tokyo in 1995.

Field of research: Catalysis, Surface science, especially environmental catalysts such as NOx reduction catalysts.

Publication list:

"Adsorption properties of oxygen and methane on Ga-ZSM-5, the origin of the selectivity of NOx reduction using methane", Takeshi TABATA, Mikako KOKITSU and Osamu OKADA, Catalysis Letters, Vol. 25, pp. 393-400 (1994).

"Relationship between methane adsorption and selective catalytic reduction of nitrogen oxide by methane on gallium and indium ion-exchanged ZSM-5", Takeshi TABATA, Mikako KOKITSU and Osamu OKADA, Applied Catalysis B-Environmental Vol.6, pp. 225-236 (1995).

"Study on catalysts of selective reduction of NOx using hydrocarbons for natural gas engines", Takeshi TABATA, Mikako KOKITSU, Hirofumi OHTSUKA, Osamu OKADA, Luigina M. F. Sabatino, and Giuseppe Bellussi, Catalysis Today, Vol.27, pp. 91-98, (1996)

"Estimation of the intracrystalline diffusion coefficient of the reactant during selective catalytic reduction of nitrogen oxide by propane on Co-ZSM-5", Takeshi TABATA and

Hirofumi OHTSUKA, Catalysis Letters, Vol. 48, pp. 203-211, (1997).

7. In view of the substantive examination report the above identified application on June 15, 1998, experiments were carried out under the directions and supervision of myself, and I beg to submit herewith an exact report thereon.

EXPERIMENT

Preparation of Cu-BEA and Ni-BEA

Cu-BEA and Ni-BEA was prepared according to the method disclosed in Japanese patent publication JP-5-220,403A(1998). In brief, 12g of Beta having a $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio of 43 was added to a solution dissolving 1.42g of copper acetate in 1 liter of water. The mixture was stirred for 3 hours at room temperature. The pH of the solution was adjusted to 11 by ammonia addition. The mixture was stirred at room temperature overnight. The solid was washed, dried at 100°C , and calcined at 550°C . The Cu loading was 3.5wt% and the Cu/Al molar ratio was 0.87.

Ni-BEA was prepared as follows. 20 grams of Beta having a $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio of 43 were added to a solution of 1.7656g of nickel acetate tetra hydrate in 400 ml of water. The mixture was heated at 60°C under stirring for 6 hours. Water was distilled under reduced pressure. The solid was dried at 110°C and calcined at 550°C . The Ni loading was 2.3wt% and the Ni/Al molar ratio was 0.62.

Catalytic activity tests

Catalytic activity of the prepared catalysts was examined in the same way as that disclosed in Japanese patent publication JP-5-220,403A(1998).

The test gas composition was 0.1% CO , 0.06% H_2 , 0.08% C_2H_6 , 4.8% O_2 , 0.1% NO , 11.9% CO_2 , 2.3% H_2O and He for balance. The test gas was passed through the catalyst at a GHSV (gas hourly space volume) of 420,000 h^{-1} , the temperature of the catalyst bed being kept at 350, 400 and 450°C , and NO_x and C_2H_6 conversions were measured. The values are listed in the left columns (fresh) of Table 1.

The catalyst was then underwent a deterioration treatment. The treatment was done by passing a gas containing 21% O_2 and 10% H_2O with the balance He for 20 h.

The treated catalysts were again subjected to the activity measurement under the same condition as described above. The NO_x and C₃H₆ conversions after the deterioration treatment are listed in the right columns (deteriorated) of Table 1.

It is obvious that Ni-BEA do not show significant NO_x conversions under the conditions disclosed in the reference even before deterioration treatment, while it shows similar C₃H₆ conversions at least before deterioration treatment. This indicates that all the listed metal other than Cu do not show similar activity and durability to Cu when they are loaded on BEA.

Table 1 Results of catalytic activity test

		fresh			deteriorated		
		Temperature (°C)			Temperature (°C)		
		350	400	450	350	400	450
Cu-BEA	NO _x conversion (%)	8	18	24	3	6	10
	C ₃ H ₆ conversion (%)	25	63	87	9	28	56
Ni-BEA	NO _x conversion (%)	3	3	4	1	1	3
	C ₃ H ₆ conversion (%)	18	46	88	0	5	21

It is declared by undersigned that all statements made herein of undersigned own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statement were made with the knowledge that willful false statement and the like so made are punishable by fine or imprisonments, or both, under 18 U.S Code 1001 and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

Date this

9th day of December, 1998

Takeshi Tabata

Takeshi TABATA

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